

1 TO WHOM IT MAY CONCERN:

2

3 BE IT KNOWN THAT WE, EMILIO CASTANO GRAFF, a  
4 citizen of the United States of America, residing in  
5 Torrance, in the County of Los Angeles, State of  
6 California, and LANCE G. HAYS, a citizen of the United  
7 States of America, residing in Placentia, in the County  
8 of Orange, State of California, have invented a new and  
9 useful improvement in

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12 CENTRIFUGE AND CASCADE FOR THE SEPARATION OF GASES

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## BACKGROUND OF THE INVENTION

This invention relates generally to separation of gases, and more particularly to improvements in method and apparatus to centrifugally separate gaseous streams.

Nearly every source of natural gas has carbon dioxide,  $\text{CO}_2$ , as an impurity. The major hydrocarbon constituent of natural gas is methane,  $\text{CH}_4$ . For some sources the percentage of  $\text{CO}_2$  may be as high as 70% while the methane component is only 30%. In order to process natural gas, the  $\text{CO}_2$  content must be reduced. Current methods of reduction include absorption in a chemical solution or use of membranes. Both of these methods are costly and require a large amount of equipment and space. For offshore production and processing of natural gas, the cost and space requirements of these conventional methods of  $\text{CO}_2$  reduction can result in an uneconomic project, reducing the recovery of the natural gas.

The possibility of using centrifugal force to separate gases was first suggested by Redig in 1895. Once isotopes were found to exist in 1913, centrifuges surged as a method of separating different isotopes by separating gaseous components. Beginning in the 1930's

1 and through the Manhattan Project in the 1940's gaseous  
2 centrifuge research was directed to the enrichment of  
3 Uranium 235 for use as nuclear fuel.

4           Although the United States abandoned the  
5 method of centrifugal separation, preferring gaseous  
6 diffusion instead, the Soviet Union and a coalition of  
7 European nations continued to research gaseous  
8 centrifuges and eventually established plants of  
9 industrial capacity using such technology to produce  
10 enriched uranium. Recently, work has been done to  
11 separate other isotopes for use in, for example, the  
12 medical field.

13           Yet the possibility of using gaseous  
14 centrifuges as disclosed herein for the separation of  
15 two completely different gases has never seriously been  
16 explored. If such a system of gaseous centrifuges were  
17 provided and operated to separate two (or more)  
18 chemically different (as in not just different isotopes  
19 of the same element), the apparatus, as provided  
20 herein, would be physically smaller and would require  
21 less resources than alternative methods which are in  
22 use today, thus providing an economically best choice.

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1                                   SUMMARY OF THE INVENTION

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3                   It is a major object of the invention to  
4 provide method and means for separating mixtures of  
5 gases into their components, in an improved and highly  
6 efficient way, or manner.

7                   A gaseous centrifuge is a relatively small,  
8 enclosed device that rotates at extremely high speeds  
9 (upwards of 30,000 RPM) which takes advantage of  
10 centrifugal force to separate a mixture of gases. Once  
11 a mixture of gases is fed into such a centrifuge, a  
12 radial concentration gradient is established in which  
13 the heavier gas is at higher concentration (than the  
14 input stream) at the periphery and the lighter gas is  
15 at a higher concentration (than the input stream) in  
16 the region closer to the axis of rotation.

17                  The partial pressure of a gaseous component  
18 in a centrifuge, assuming solid body rotation is given  
19 from the Maxwell-Boltzmann Distribution Laws as:

20  
21                                   
$$p_i(r) = p_i(0) \exp [M_i (\Omega r)^2 / 2RT]$$

22       where:

23                    $p_i$  = partial pressure of component 1 at  
24                   location ()

25                    $M_i$  = molecular weight of component 1

1             $\Omega$  = angular velocity

2             $r$  = radius

3             $R$  = gas constant

4             $T$  = temperature

5

6    Comparing the concentration of two gaseous components  
7    gives:

8                             $\alpha = \exp[(M_2 - M_1)(\Omega r)^2 / 2RT]$

9    where:

10

11             $\alpha$  = ratio of concentration of component 2 to  
12            component 1

13             $M_2$  = molecular weight of component 2

14             $M_1$  = molecular weight of component 1

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16            For separation of isotopes such as uranium  
17    235 and uranium 238 the molecular weight difference is  
18    only 3 units resulting in a relatively small  
19    concentration factor and a huge number of concentration  
20    stages required to effect a substantial concentration.  
21    However, for mixtures of carbon dioxide and methane,  
22    the molecular weight difference is

23

24                             $M_2 - M_1 = 44 - 16 = 28$

1           This produces a large concentration ratio  
2 compared to typical isotope separations. Consider a  
3 speed of 3000 radians/second, a cylinder radius of 10  
4 cm and a temperature of 300 °K. For uranium 235 and  
5 uranium 238 separation, the concentration ratio is:

6  
7            $\alpha_1 = \exp[(3)(300)^2/(2)(8314)(300)] = 1.056$

8  
9 For CO<sub>2</sub> and CH<sub>4</sub> the concentration ratio is:

10  
11            $\alpha_1 = \exp[(28)(300)^2/(2)(8314)(300)] = 1.657$

12           Thus, an unexpected result as disclosed  
13 herein is found in applying a centrifuge to separate  
14 carbon dioxide from methane in that an extraordinary  
15 increase in concentration can be accomplished compared  
16 to isotopic separation.

17           Accordingly, another major object is to  
18 provide a gas centrifuge means operating to separate  
19 gases of differing chemical composition and molecular  
20 weight by a centrifugal force field. Typically, and in  
21 accordance with a further feature of the invention,  
22 carbon dioxide is separated from methane by an improved  
23 method employing a centrifugal force field.

24           Another object is to provide a multiplicity

1 of centrifuge means as defined in claim 1, arranged  
2 such that the separated streams of gases are further  
3 concentrated by introducing them into successive of the  
4 gas centrifuge means.

5 An additional object is to provide a gas  
6 processing system utilizing centrifugal force for the  
7 separation of light gases from heavy gases, liquids  
8 from gases, light liquids from heavy liquids and solids  
9 from liquids and gases.

10 An additional object is to provide a gas  
11 centrifuge apparatus comprising, in combination:

12 a) a hollow shaft to pass and introduce a  
13 gas mixture into a rotating cylinder,

14 b) said cylinder having axial vanes to  
15 cause the gas mixture to rotate with the same angular  
16 speed as the cylinder,

17 c) a radial passage connected to the  
18 periphery of the cylinder to receive and pressurize a  
19 produced and concentrated heavier gas stream,

20 d) a nozzle connected to the passage to  
21 convert the pressure of the heavier gas stream to  
22 velocity adding a torque to the cylinder, and

23 e) an opening in the hollow shaft to  
24 receive and remove a produced and concentrated lighter  
25 gas stream from the cylinder.

1           A yet further object is to provide an  
2 improved centrifuge apparatus operating in the manner  
3 referred to, and incorporating:

4           a)    a nozzle accelerating a gas mixture and  
5 introducing it into a rotating cylinder, adding torque  
6 to the cylinder,

7           b)    the cylinder having vanes to receive  
8 torque from the gas and causing the gas to rotate with  
9 the same angular speed as the cylinder,

10          c)    a radial passage connected to the  
11 periphery of the cylinder operating to pressurize a  
12 produced and concentrated heavier gas stream,

13          d)    a nozzle connected to the passage and  
14 operating to convert the pressure of the heavier gas  
15 stream to velocity, adding torque to the cylinder,

16          e)    an open scoop oriented perpendicular to  
17 the direction of rotation operating to remove a  
18 produced and concentrated lighter gas from the  
19 cylinder, and

20          f)    a passage contoured and operating to  
21 recover the velocity head of the concentrated lighter  
22 gas as pressure.

23           A yet further object is to provide an  
24 improved centrifuge apparatus operating in the manner  
25 referred to, and incorporating;



1           a)    a nozzle accelerating a gas mixture and  
2    introducing it into a rotating cylinder, adding torque  
3    to the cylinder,

4           b)    the cylinder having vanes to receive  
5    torque from the gas and causing the gas to rotate with  
6    the same angular speed as the cylinder,

7           c)    a radial passage connected to the  
8    periphery of the cylinder operating to pressurize a  
9    produced and concentrated heavier gas stream,

10          d)    a nozzle connected to the passage and  
11    operating to convert the pressure of the heavier gas  
12    stream to velocity, adding torque to the cylinder,

13          e)    a radial passage connected to the  
14    periphery of the cylinder, extending radially inward  
15    such that its inlet is at the region of concentration  
16    of the lighter gas and operating to pressure a produced  
17    and concentrated lighter gas stream,

18          f)    a nozzle connected to the radial passage  
19    and operating to convert the pressure of the  
20    concentrated lighter gas to velocity, adding torque to  
21    the cylinder.

22                These and other objects and advantages of the  
23    invention, as well as the details of an illustrative  
24    embodiment, will be more fully understood from the  
25    following specification and drawings, in which:

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## DRAWING DESCRIPTION

Fig. 1 is a section taken through one form of improved gas separation apparatus embodying the invention; and Figs. 1a and 1b are sections taken on lines 1a-1a and 1b-1b shown in Fig. 1;

Fig. 2 is a section taken through another form of improved gas separation apparatus embodying the invention;

Fig. 3 is a centrifuge cascade system block diagram;

Fig. 4 is a graph showing how the number of centrifuge stages increases methane concentration where methane is being centrifugally separated from carbon dioxide;

Fig. 5 is a section taken through another form of improved gas separation apparatus embodying the invention; and

Fig. 6 is a showing of a modification.

## DETAILED DESCRIPTION

Fig. 1 shows a centrifuge that can be used to decrease the concentration of  $\text{CO}_2$  in a gas mixture without any chemical absorption or membranes.

1           A mixture of methane,  $\text{CO}_2$ , and any other gas  
2 species is introduced at 1 to the centrifuge 100, via a  
3 hollow shaft 2. The shaft is supported by bearings as  
4 at 12 and the gas is introduced to the hollow shaft  
5 through a fixed seal assembly associated with 12. The  
6 gas flows axially into the centrifuge from the shaft  
7 through side opening 3.

8           The gas in the centrifuge interior 40 is  
9 subjected to the centrifugal force produced by the  
10 rotation of the centrifuge. Rotation of the gas is  
11 caused by axial vanes 40a attached to the centrifuge  
12 shaft. Such rotation can be produced either by  
13 applying a torque to the shaft 2, or by causing a  
14 pressure drop across a nozzle 8, which produces a  
15 reaction force from outflow of the gas, within 40.

16           The heavier carbon dioxide gas ( $\text{CO}_2$ ,  
17 molecular weight = 44, and other heavier gases such as  
18  $\text{H}_2\text{S}$ , molecular weight = 34) is concentrated at the  
19 centrifuge outer radius zone 4, near outer cylindrical  
20 wall 104. The lighter methane gas ( $\text{CH}_4$ , molecular  
21 weight = 16) is concentrated at the inner radius zone 5  
22 near inner cylindrical wall 105. The carbon dioxide  
23 rich gas is removed through a passage 7 communicating  
24 with zone 4, and reaction nozzle 8, at the periphery of  
25 the centrifuge rotor part 18. The carbon dioxide is  
26 isolated from the lower pressure gas in zones 15, 16,

1 and 17, surrounding the centrifuge rotor by annular  
2 seals, 13 and 14, between 18 and bore 106 of housing  
3 107. The concentrated carbon dioxide is removed  
4 through a volute 9, discharging at 110.

5 The enriched methane is removed through a  
6 port, 10, in the hollow shaft 2, and flows at 11 to  
7 another part 110 of the process. Wall 6 in the shaft  
8 separates flows 1 and 11. Auxiliary means to rotate  
9 the shaft is shown at 111.

10 Fig. 2 shows another centrifuge which can be  
11 used to decrease the concentration of carbon dioxide in  
12 a gas mixture.

13 A gas mixture 1', enters the centrifuge 100'  
14 via wall 110' and flows to a nozzle 2', which is  
15 oriented in a generally tangential direction to a  
16 cylindrical rotor 18'. The gas mixture is expanded in  
17 the nozzle to a high exit velocity at 3', in a  
18 direction generally tangential to the rotor. The gas  
19 flows through axial vanes 17', with turbine effect,  
20 which support the rotor from a shaft 16'. Nozzle 2' is  
21 radially offset relative to rotary shaft 16'. The  
22 rotor acquires the circumferential velocity component  
23 of the entering gas.

24 The heavier carbon dioxide is concentrated by  
25 the centrifugal force at centrifuge outer radius zone  
26 4', near outer wall of the rotor 104. The lighter

1 methane is concentrated at the inner radius zone 5'  
2 near the surface of shaft 16' of the rotor. The  
3 concentrated carbon dioxide stream flows through outlet  
4 passage 7' increasing its pressure. The flow is then  
5 accelerated through a nozzle 8, adding more torque to  
6 the rotor to overcome windage and friction losses. The  
7 concentrated carbon dioxide stream is removed through a  
8 volute 9', discharging at 209'.

9           The concentrated methane stream flows into an  
10 outlet scoop 10', which faces in generally tangential  
11 relation to the circumferential flow direction to  
12 remove a produced and concentrated lighter gas such as  
13 methane, from the cylinder. The velocity is converted  
14 to pressure by the passage 11', which has an increasing  
15 flow area within wall 111' to diffuse the velocity and  
16 recover the velocity head as increased pressure at 12',  
17 and delivered at 300 to process 301. The concentrated  
18 methane is removed through another volute 12' at the  
19 outer side or end of 111'.

20           The rotor is supported by annular bearings  
21 13' located between the shaft 16 and bores in end walls  
22 110' and 111'. If sufficient pressure drop is  
23 available between 1' and 3', the shaft may be totally  
24 enclosed; otherwise, a seal is incorporated in the  
25 structure 13', and a power source 301' is provided to  
26 rotate the centrifuge at desired speed.

1           The pressure within the rotor 18' is isolated  
2 by annular seals 14 and 15 from the low pressure on the  
3 outer side 19' of the rotor, which is required to  
4 minimize frictional losses at the high speed of the  
5 rotor. The concentrated CO<sub>2</sub> in the volute 9', is  
6 isolated from the pressure within the rotor 18', and  
7 the pressure at zone 19' surrounding the rotor, by  
8 seals 14' and 15'.

9           Fig. 5 shows another centrifuge which can be  
10 used to decrease the concentration of carbon dioxide in  
11 a gas mixture.

12           A gas mixture 1', enters the centrifuge 104''  
13 via wall 110' and flows to a nozzle 2', which is  
14 oriented in a generally tangential direction relative  
15 to a cylindrical rotor 18'. The gas mixture is  
16 expanded in the nozzle to a high exit velocity at 3'',  
17 in a direction generally tangential to the cylindrical  
18 rotor. The gas flows through axial vanes 17'', with  
19 turbine effect, which support the rotor from a shaft  
20 16'. Nozzle 2' is radially offset relative to rotary  
21 shaft 16'. The rotor acquires the circumferential  
22 velocity component of the entering gas.

23           The heavier carbon dioxide is concentrated by  
24 the centrifugal force at centrifuge outer radius zone  
25 4', near outer wall 104'' of the rotor. The lighter  
26 methane is concentrated at the inner radius zone 5'

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1 near the surface of shaft 16' of the rotor. The  
2 concentrated heavier carbon dioxide stream flows  
3 through outlet passage 7'', increasing its pressure.  
4 The flow is then accelerated through a nozzle 8'',  
5 adding more torque to the rotor to overcome windage and  
6 friction losses. The concentrated carbon dioxide  
7 stream is removed through a volute 9', discharging at  
8 209''.

9 The concentrated methane stream flows into  
10 another outlet passage 10'', whose inlet 10''' is  
11 located radially inward at the radial location 5''  
12 where the lighter gas is concentrated. The  
13 concentrated methane stream flows through the outlet  
14 passage 10'' increasing it's pressure. The flow is  
15 then accelerated through a nozzle 8''' adding more  
16 torque to the rotor to overcome windage and friction  
17 losses. The concentrated methane is removed through  
18 another volute 9''' discharging at 209''.

19 The rotor is supported by annular bearings  
20 13'' located between the shaft 16'' and bores in end  
21 walls 110'' and 111''.

22 The pressure within the rotor 18'' is  
23 isolated by annular seals 14'' and 15'' from the low  
24 pressure on the outer side 19'' of the rotor, which is  
25 required to minimize frictional losses at the high  
26 speed of the rotor. Such seals seal off between 18''

1 and wall 110a''. The concentrated CO<sub>2</sub> in the volute  
2 9'', is isolated from the pressure within the rotor  
3 18'' and the pressure at zone 19'' surrounding the  
4 rotor, by seals 14'' and 15''.

5 To further concentrate the carbon dioxide  
6 stream and the methane stream, the flows at 9' and 12'  
7 leaving the centrifuge from Fig. 5, can be introduced  
8 to additional like centrifuges, i.e. a ''cascade'' of  
9 centrifuges.. The cascade provides a method of  
10 connecting many centrifuges together so as to amplify  
11 the separation capacity and flow rate of a single unit.

12 The cascade is typically comprised of a  
13 number of stages, the size of each stage being defined  
14 by the amount of flow that must go through the cascade.  
15 The amount of flow required is directly related to the  
16 desired flow of the product (the stream comprised  
17 mostly of the lighter gas) and its concentration. The  
18 desired concentration, in turn, determines the number  
19 of stages necessary. The product delivery end of the  
20 cascade is called the ''top'' while the waste end is  
21 called the ''bottom''.

22 The cascade is divided into two sections, the  
23 ''stripper'' and the ''enricher''. The enricher  
24 section is that between the feed point (where the  
25 mixture comes in) and the top of the cascade, while the  
26 stripper section is the section below the feed point.



1 These sections are called stripper and enricher because  
2 the stripper can be thought of as concentrating the  
3 waste (heavier) gas, while the enricher concentrates  
4 the product (lighter) gas.

5 All the stages except the top, bottom, and  
6 the first enricher stage have equivalent connections.  
7 The feed is comprised of the waste of the stage above  
8 and the product of the stage below. The feed of the  
9 top stage is only the product of the stage below it,  
10 while the feed of the bottom stage is only the waste of  
11 the one above it. The feed at the first enricher stage  
12 is comprised of the product from the stage below it,  
13 the waste from the stage above it, and the feed into  
14 the cascade.

15 To avoid mixing, and therefore to make the  
16 cascade as efficient as possible, each stage has a  
17 different proportion of its output that is selected as  
18 the product and the waste. This proportion is called  
19 the cut and is directly related to the desired product  
20 flow, the concentrations of the outputs, and the  
21 separation power of the centrifuge.

22 Fig. 3 shows a cascade arrangement of six  
23 centrifuges. More or less can be used with the same  
24 principles. A flow mixture of carbon dioxide and  
25 methane and/or other gases 207, enters a centrifuge  
26 201. A carbon dioxide concentrated stream 210, from

1 another centrifuge 202, is also introduced to the first  
2 centrifuge 201. An enriched methane stream 218, from  
3 another centrifuge 203, is also introduced to the first  
4 centrifuge 201. The composition of the carbon dioxide  
5 concentrated stream 210, from centrifuge 202, and the  
6 composition of the enriched methane stream 218 from  
7 centrifuge 203, is made equal or nearly equal to the  
8 composition of the initial stream 207.

9           The enriched methane stream 208, from the  
10 first centrifuge 201, enters the second centrifuge 202.  
11 A carbon dioxide concentrated stream 212, from another  
12 centrifuge 204, having an equal or nearly equal  
13 composition as stream 208, is also introduced to  
14 centrifuge 202. The concentrated carbon dioxide stream  
15 210, leaving centrifuge 202, is introduced to the first  
16 centrifuge 201. The enriched methane stream 211,  
17 leaving centrifuge 202 is introduced to centrifuge 204.

18           The enriched methane stream 213, leaving  
19 centrifuge 204, is the product stream of enriched  
20 methane, having a minimum amount of carbon dioxide.  
21 The concentrated carbon dioxide waste stream 212, from  
22 centrifuge 204 is introduced to centrifuge 202 for  
23 further concentration.

24           The concentration of the waste carbon dioxide  
25 stream 209, from the first centrifuge 201, is increased  
26 by introducing the stream into another centrifuge 203.

1 The enriched methane stream 218, from centrifuge 203 is  
2 fed to the first centrifuge 201. The concentrated  
3 carbon dioxide stream 213, leaving centrifuge 203, is  
4 introduced to another centrifuge 205, for further  
5 concentration.

6 The enriched methane stream 214, is  
7 introduced to centrifuge 203 for further enrichment.  
8 The concentrated carbon dioxide stream 215, is  
9 introduced to another centrifuge 206, for further  
10 concentration.

11 The enriched methane stream 216, from  
12 centrifuge 206 is introduced to centrifuge 205 for  
13 further enrichment. The concentrated carbon dioxide  
14 waste stream 217, from centrifuge 206 is the final  
15 ''waste'' stream and flows to that part of the process  
16 where it is used or disposed of.

17 The enrichment of methane in the product  
18 stream and the concentration of carbon dioxide in the  
19 waste stream are shown in graph form in Fig. 4 for a  
20 cascade using centrifuges operating at 48,000 rpm, each  
21 having a radius of 8 centimeters. The initial  
22 concentration is 30% methane (by mole) and 70% carbon  
23 dioxide.

24 The concentration of methane increases from  
25 30% to 86% with 6 stages. Increasing the number of  
26 stages to 16 results in virtually complete separation.

1           The use of centrifugal forces in the gas  
2 centrifuge can be combined with centrifugal forces in  
3 other devices to produce a gas processing system which  
4 is very compact and which utilizes the energy in high  
5 pressure gas sources to reduce energy consumption.

6           Fig. 6 illustrates a centrifugal gas  
7 processing system.

8           Untreated gas 101 enters a Three-Phase Rotary  
9 Separator 102. The Three-Phase Rotary Separator  
10 separates solids 103, free water 104, and free  
11 hydrocarbon liquids 105 from the gas.

12           The saturated gas 106 flows from the Three-  
13 Phase Separator into the Integral Separator 112. The  
14 Integral Separator is a centrifugal gas-liquid  
15 separator, which derives all or part of the rotational  
16 energy required from the gas pressure letdown. The  
17 functions of the Integral Separator are:

- 18           • To lower the temperature of the gas  
19           stream by near isentropic expansion in  
20           order to condense out natural gas  
21           liquids and water, if present.
- 22           • Separation of the liquids from the gas  
23           using a rotating separation surface  
24           driven by the fluid energy.

- Re-compression of the separated gas with a radial diffuser to decrease the dewpoint (dehydration) and remove the kinetic energy from the flow.

Methanol or another absorbent 107 can be injected in the Integral Separator nozzles and separated for re-use 108. The separated hydrocarbon liquids 109 are collected for use. The waste products 110 from the absorbent treating system are collected for disposal or further treatment.

The dry separated gas 111 flows into the gas centrifuge cascade 113. The gas centrifuge separates the heavier gases such as CO<sub>2</sub>, H<sub>2</sub>S and sulfur compounds from the much lighter methane. The large molecular weight difference results in higher concentration relative to other gas centrifuge applications. Fluid and/or shaft energy from the Integral Separator is used to supply or augment the power required for the gas centrifuge rotation. a motor drive 114 can provide the balance if required. In the event of excess shaft energy, such as can be achieved for high gas pressures, a generator can be used to generate power for other parts of the facility.

1           The dry methane 115 is delivered to the  
2 pipeline. The CO<sub>2</sub>, H<sub>2</sub>S and sulfur compounds 116 are  
3 delivered for re-injection or treatment.

4           The system's primary benefits are:

- 5           • Compact size resulting from centrifugal  
6 processing.
- 7           • Maximization of the recovery of natural  
8 gas liquids (NGL).
- 9           • Low energy consumption (or energy  
10 production).
- 11          • Reduction of the balance of plant.
- 12          • Reduction of chemical requirements.
- 13          • Portability.
- 14          • Marinization potential for subsea  
15 application.